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## **THERMAL DIFFUSION OF WATER VAPOUR IN POROUS MATERIALS: FACT OR FICTION ?**

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### **ABSTRACT**

The reliable evaluation of moisture transfer in porous materials is essential in many engineering applications, among which building science. One key aspect is a correct description of moisture flow phenomena and their transport potentials. While different issues can be debated in that respect, thermal diffusion of water vapour in porous materials – diffusion driven by temperature gradients – currently stands out, due to the contradictory findings on the topic. Thermal diffusion proponents uphold that, complementary to vapour pressure gradients, temperature gradients equally yield substantial diffusion. Thermal diffusion opponents, on the other hand, assert that these thermal transports are negligibly small.

This paper resolves that contradiction. A critical analysis of the investigations supporting the occurrence of thermal diffusion reveals that all are flawed. A correct reinterpretation of all measurements allows concluding that no consistent nor significant thermal diffusion can be observed. This brings these investigations in line with their earlier opponents. This conclusion also agrees with thermodynamics, which confirms the actual existence of thermal diffusion, but also indicates its negligible magnitude. It can in conclusion be stated that thermal diffusion is of no importance for building science applications, leaving vapour pressure as the sole significant transport potential for the diffusion of water vapour in porous materials.

### **KEYWORDS**

Vapour diffusion, Fick's law, Soret effect, thermal diffusion, vapour pressure

## NOMENCLATURE

c	specific heat [J/kg·K]
C	mass fraction [kg/kg]
D	vapour diffusivity for vapour concentration gradients [m <sup>2</sup> /s]
D'	vapour diffusivity for temperature gradients [m <sup>2</sup> /K·s]
h	heat of vaporisation [J/kg]
j	diffusion flow [kg/m <sup>2</sup> ·s]
L <sub>vv</sub>	phenomenological coefficient [kg·K/J·m·s]
L <sub>vq</sub>	phenomenological coefficient [kg·K/m·s]
P	total pressure [Pa]
p	partial pressure [Pa]
q	heat flow [W/m <sup>2</sup> ]
R	gas constant [J/kg·K]
RH	relative humidity [%]
T	temperature [K]

### *Greek symbols*

α	thermal diffusion factor [-]
δ <sub>p</sub>	permeability for vapour pressure gradients [kg/m·s·Pa]
δ <sub>T</sub>	permeability for temperature gradients [kg/m·s·K]
ε	thermal diffusion factor [
μ	chemical potential [J/kg]
σ	entropy production [W/kg·K]

### *Subscripts/superscripts*

a	air
act	actual value
avg	at average temperature
mea	measured value
p	due to vapour pressure gradients
q	heat
T	due to temperature gradients
v	vapour
w	water
wet	during evaporation
dry	after evaporation
+	at warm chamber temperature
-	at cold chamber temperature
⊗	reference value

## 1. INTRODUCTION

Moisture transfer in porous materials intervenes in many processes, be it at the macro-scale of nuclear waste storage [1] or at the microscale of cement particle hydration [2]. Transfer of moisture, and the chemicals dissolved therein, similarly plays a main role in the durability and sustainability of built structures, and in the health and comfort of building occupants. The corrosion of concrete reinforcement bars, due to chloride ingress via the pore water, exemplifies potential durability issues [3], whereas mould formation on building components, caused by excessive interior relative humidity levels, illustrates possible health issues [4]. Moisture is often a crucial determinant when assessing built structures' durability and sustainability or building occupants' health and comfort. Hence, for the correct design of new structures or for the remediation of defective existing ones, a reliable evaluation of moisture transport in porous building materials is crucial. Many hydrological, agricultural and environmental applications, and various other engineering areas, equally need a dependable assessment of moisture transfer. One key aspect here is the correct description of moisture flow phenomena and their transport potentials. While many issues can be debated in that respect, thermal diffusion of water vapour in porous materials – diffusion driven by temperature gradients – currently stands out due the contradictory findings on the topic.

The proponents of thermal diffusion [5-9] uphold that, complementary to vapour pressure gradients, temperature gradients equally yield sizeable diffusion. The opponents of thermal diffusion [9-12], on the other hand, assert that such thermal transport is negligibly small. The thermal diffusion proponents thus suggest that vapour pressure and temperature are both significant transport potentials for diffusion:

$$\dot{j}_v = \dot{j}_p + \dot{j}_T = -\delta_p \nabla p_v - \delta_T \nabla T \quad (1)$$

where  $\dot{j}_v$ ,  $\dot{j}_p$  and  $\dot{j}_T$  [kg/m<sup>2</sup>·s] are global, standard and thermal vapour flow,  $p_v$  [Pa] and  $T$  [K] vapour pressure and temperature, and  $\delta_p$  [kg/m·s·Pa] and  $\delta_T$  [kg/m·s·K] are permeabilities for  $p_v$  and  $T$  gradients. The thermal diffusion opponents, in contrast, advocate that vapour pressure is the sole significant transport potential for diffusion.

Most research on thermal diffusion in porous materials [5-12] is experimental, and all investigations apply the same measurement concept. The diffusion flow through a single layer of material under combined gradients of vapour pressure and temperature is measured, and compared to the respective isothermal diffusion flow. Several of these studies [5-9] support 'significant thermal diffusion'. They all find positive thermal diffusion – from high to low temperature –, with thermal permeabilities  $\delta_T$  one or two orders of magnitude larger than the respective standard permeabilities  $\delta_p$ . Even while in most building science applications vapour pressure gradients are usually far larger than temperature gradients, such thermal permeabilities would have a considerable impact. On the other hand, multiple other studies [9-12] use similar principles and conditions, but find no consistent nor significant evidence for thermal diffusion. This paper aims at reconciling these contradictory findings on thermal diffusion and arriving at a convergent conclusion on thermal diffusion of water vapour in porous materials.

It should be noted that this paper focuses only on the diffusion of water vapour and not on moisture (liquid and vapour) transfer in general. While liquid transfer is sometimes termed diffusion as well, it is now generally accepted that this process is mainly driven by gradients in capillary pressure, and not by gradients in water concentration [9]-[14]. The term 'diffusion' for general moisture transfer should hence be considered outdated. Whereas vapour diffusion is of course surpassed in magnitude by capillary transport of liquid, or advective transport of vapour, it still forms a notable moisture transfer mechanism. Water vapour diffusion is essential in the assessment of interstitial condensation in building parts [15], moisture buffering by interior enclosures [16], hygrothermal comfort of clothing [17], evaporative cooling of roofs [18], ....

This paper aims at reconciling the contradictory findings on thermal diffusion. Introductorily, the studies finding no evidence for thermal diffusion [9-12] are reanalysed, and their conclusions are confirmed. In the major section of the paper, the studies supporting thermal diffusion [5-9] are dissected, and they are demonstrated to be flawed. A corrected reinterpretation demonstrates that they neither find consistent nor significant evidence for thermal diffusion. Lastly, the experimental findings are complemented by a thermodynamic analysis, which will finally permit to reach the convergent conclusion that the thermal diffusion of water vapour in porous materials is insignificant.

## **2. INSIGNIFICANT THERMAL DIFFUSION**

With multiple studies [5-9] asserting important positive thermal diffusion as backdrop, several other studies [9-12] did not detect consistent nor significant evidence for thermal diffusion. The latter are reanalysed here first, to confirm their original findings and conclusions.

### **2.1 Galbraith et al., 1998 [9]**

#### **2.1.1 Authors' measurements**

Galbraith et al. performed non-isothermal vapour diffusion measurements on two materials: 12 mm particle board and 25 mm polystyrene insulation. The common cup set-up was used, with a humidity regulator in the cup and the cup placed in a climate chamber. The cup was suspended though in a thermostatic bath at 15 °C, while 25 °C was maintained in the chamber. A measurement overview can be found in Appendix 1.

#### **2.1.2 Authors' observations**

Comparison of non-isothermal results with isothermal cup measurements permitted determining the thermal diffusion flows  $j_T$ . The authors inferred that "for the particle board the estimated thermal diffusion rates are extremely small. The largest is indicated as only 5% of the total moisture flux which is considerably less than the standard deviation of the measurements of  $j$ . In addition no definite trend is obvious in that the sign of the thermal flow is not constant." The authors further stated that "for the polystyrene  $j_T$  has a consistent sign and it constitutes a larger percentage of the total flux. However, only at one of the test conditions does its magnitude exceed the standard deviation of  $j$ ." It was thus concluded that "these results would seem to provide no evidence of any significant thermal diffusion".

#### **2.1.3 Additional analysis**

Figure 1 compares the non-isothermal vapour flows with the expected isothermal flows, and puts them into perspective by inserting the standard deviation on each of the non-isothermal flows and the overall standard deviation on the isothermal flows. That value is derived as follows. The given standard deviations on the permeability (Table 1 in [9]) are converted to standard deviations on the flows. These are only weakly linked to the flows' magnitude and are thus aggregated into one standard deviation on the expected isothermal flows. Vapour diffusion flows and vapour pressure differences are assumed positive if from the warm to the cold chamber. It is clear that most differences between non-isothermal and isothermal flows are below standard deviation. Moreover, while the particle board shows primarily positive differences (from warm to cold), the polystyrene insulation gives mainly negative differences. It can only be concluded that no consistent nor significant thermal diffusion is detected.

## **2.2 Thomas, 1999 [10]**

### **2.2.1 Authors' measurements**

Thomas performed measurements on 6.35 mm oriented strand board. He used a two-chamber set-up, with the sample forming the horizontal boundary between the bottom and top chamber, with each chamber maintained at a specific temperature and relative humidity by air conditioning systems.

The author set out to establish the true transport potential for vapour diffusion, selecting from: moisture concentration, vapour pressure, vapour pressure/temperature, activated moisture content, chemical potential. Only a true potential would yield zero vapour flow when its values are the same in both chambers. A variety of conditions was used, with temperatures ranging from 5 °C to 35 °C. An overview is given in Appendix 1.

### **2.2.2 Authors' observations**

The author deduced that "diffusion under non-isothermal conditions is governed by the gradients of the vapour pressure. This observation is based on the criterion that moisture transfer must cease if the diffusion driving force is the same on both sides of the test material."

### **2.2.3 Additional analysis**

Figure 2 compares the non-isothermal vapour flows with the expected isothermal flows, with insertion of the standard deviations on the measurements. Vapour diffusion flows and vapour pressure differences are assumed positive when from the warm to the cold chamber. Figure 2 clearly shows that deviations are mostly very small and inconsistent in sign. Furthermore, no trend with the imposed temperature difference (10 K and 5 K) can be distinguished. No consistent nor significant thermal diffusion can thus be detected.

It should furthermore be considered that the two 'negative flow' measurements (bottom left in Figure 2) impose nearly the same conditions, only differing by the position of the warm and cold chambers. Identical conditions should result in identical flows: it can be seen though that with a warm chamber at the bottom a lower flow is obtained than with a warm chamber at the top. The big deviation suggests that the given standard deviation actually underestimates the systematic error on the measurements.

## **2.3 Baker et al., 2009 [12]**

### **2.3.1 Authors' measurements**

Baker et al. performed measurements on 9 different materials. They equally applied a two-chamber set-up, both climatized by air conditioning systems, however with a vertical sample in between. The core of their measurements targeted 'zero vapour pressure gradient' tests, nullifying the vapour pressure difference between the two chambers. Temperatures ranged from 12 °C to 29 °C.

### **2.3.2 Authors' observations**

Lack of stringent climate control only allowed approximations of 'zero vapour pressure gradient'. Instead, the authors measured several flows at small vapour pressure differences (up to 300 Pa), and the expected intercepts at '0 Pa vapour pressure difference' were derived from linear regression. Their results for phenolic foam are exemplarily given in Figure 3. For all materials investigated it was shown that these '0 Pa'-flows did not deviate significantly from zero, as also illustrated in Figure 3. Moreover, the signs of the deviations are, as before, not consistent. From these observations, the authors concluded that thermal diffusion "has no measurable significance for moisture diffusion in construction materials. Generally, non-isothermal tests show that the vapour pressure gradient is the critical driving potential for moisture transfer."

### 2.3.3 Additional analysis

Introductorily, the authors also performed 'constant relative humidity' measurements, in which they imposed the same relative humidity in the warm and the cold chamber. The resulting permeabilities were then compared to cup measurements, to confirm the reliability of the two-chamber set-up. It should be remarked here that such confrontation actually assumes 'no significant thermal diffusion' as premise; but that has no consequences for the analysis below.

The relative differences between the non-isothermal and isothermal vapour permeability are shown in Figure 4, in reference to the temperature difference between the chambers. It can be observed that rather large deviations occur, both positive and negative. These are an indication of inaccuracies in the measurements, and not of thermal diffusion. Moreover no real trend with the applied temperature difference can be observed. No consistent nor significant thermal diffusion can thus be detected. No further analysis of the 'zero vapour pressure gradient' tests is performed here as the authors' study is quite satisfactory.

### 2.4 Glass, 2007 [11]

Glass measured on 11 mm exterior grade plywood. He equally applied a two-chamber set-up with a vertical sample, but with relative humidities imposed by salt solutions. The chambers were kept at about 21 °C and 27 °C. Only three humidity conditions were imposed, all nearly nullifying the vapour pressure difference between the chambers. The author concluded that his "experiments with conditions of nearly constant water vapour pressure in the presence of thermal gradient show small moisture fluxes from the cold side to the warm side that might have been caused by water vapour pressure gradients within the experimental error." No consistent nor significant thermal diffusion can hence be detected. Given the restricted set of measurements (3 measurements, all with very small vapour pressure differences), no further analysis is performed here.

## 3. SIGNIFICANT THERMAL DIFFUSION

While the studies above [9-12] found no consistent nor significant evidence of thermal diffusion, other studies [5-9] did present such evidence, even while applying the same principles and conditions. Below however the flaws in each of those are demonstrated, and corrected where possible. It will then become clear that their initial support of thermal diffusion no longer holds.

### 3.1 Kumaran 1987 [5]

#### 3.1.1 Authors' measurements

Kumaran used a heat flow apparatus to indirectly measure vapour diffusion in glass fibre insulation. A sample was wetted at one surface and sealed in a plastic wrap. With the wet side of the sample on the warm plate, the liquid evaporates and diffuses to the cold plate, where it condenses again. Measurements of sensible and latent heat flows during and after evaporation allow determination of the vapour diffusion flow.

In the most cited publication [5], Kumaran measures on glass fibre with thickness 8.55 cm and density 17 kg/m<sup>3</sup>, with temperature differences ranging from 28 K to 39 K, and on glass fibre with thickness 5.47 cm and density 30 kg/m<sup>3</sup>, with temperature differences between 21 K and 39 K. An overview is given in Appendix 1.

#### 3.1.2 Authors' observations

Kumaran suggested a simplified representation of the phase change and diffusion process to translate heat flows into diffusion flows. He assumed that the liquid evaporated at the warm side, wholly diffused to the cold side, to finally cool down and condense. It was stated that the vapour diffusion can then be calculated as:

$$j_v = (q_{\text{wet}} - q_{\text{dry}}) / (h_{w \rightarrow v, \text{avg}} + c_w (T_+ - T_-)) \quad (2)$$

where  $q$  [ $\text{W/m}^2$ ] is the heat flow,  $h_{w \rightarrow v}$  [ $\text{J/kg}$ ] the heat of vaporisation of water,  $c_w$  [ $\text{J/kg}\cdot\text{K}$ ] the specific heat of water, subscripts wet/dry point to heat flows during and after evaporation, and subscripts +/-/avg indicate the temperatures of the warm and cold chamber and their average respectively.

Linear regression of the obtained vapour flows with the imposed vapour pressure differences yielded a positive non-zero intercept, an indication of a diffusion flow not attributable to vapour pressure gradients. These observations led the author to conclude that “even in the absence of a vapour pressure gradient, moisture transfer may exist. A driving force other than vapour pressure is also responsible for the moisture transport. The obvious choice for the second potential in the present experimental investigation is the temperature.” An alternative linear regression, applying vapour pressure and temperature as transport potentials finally gave the thermal permeabilities  $\delta_T$ . These were positive, a sign of diffusion from high to low temperature, and in magnitude 17 and 36 times bigger than their  $\delta_p$  (for 17 and 30  $\text{kg/m}^3$  glass fibre respectively).

### 3.1.3 Additional analysis

The analysis discussed above [5] is the first in a series of heat flow experiments on wet glass fibre insulation [24-26]. In these, measurements on glass fibre samples with various thicknesses (2.5 to 15.4 cm) and densities (45 to 117  $\text{kg/m}^3$ ) are reported. First in [26], a more refined method to derive the vapour flows is presented:

$$j_{v,+} = (q_{\text{wet},+} - q_{\text{dry}}) / (h_{w \rightarrow v,+}) \quad j_{v,-} = (q_{\text{wet},-} - q_{\text{dry}}) / (h_{w \rightarrow v,-} + c_v (T_+ - T_-)) \quad (3)$$

Eq. (3) more correctly accounts for the enthalpy changes at both plates, as well as for the small differences in the ‘wet’ heat flows at the warm and cold plate. While this simplified model neglects all condensation of water vapour inside the insulation – a consequence of the dropping temperature in the samples –, it reliably quantifies vapour diffusion flows. This has been verified by comparison of Eq. (3) with a numerical model including internal condensation in the samples [7].

The deviations with Eq. (2) are however below 5 %, and do not significantly modify the above conclusions. All reported results have nevertheless been reprocessed with Eq. (3). If only an average  $q_{\text{wet}}$  was given, it was used as both  $q_{\text{wet},+}$  and  $q_{\text{wet},-}$ . An average of the resultant  $j_{v,+}$  and  $j_{v,-}$  was finally accepted as the diffusion flow through the sample. An overview of all results can be found in Appendix 1.

Figure 5 presents the results from the in total 17 measurement series, showing the vapour flow in function of the vapour pressure difference. For ease of presentation, each measurement series has been recalculated to a sample with unit thickness. Additionally, linear regressions through these measurements are presented. The positive  $\delta_T$ ’s as reported by Kumaran [5] are essentially based on a positive intercept at a 0 Pa vapour pressure difference (seen in light of the convention made here: vapour flows are positive if from warm to cold). In Figure 5 it can be seen that the sign of the intercepts is not consistent: positive and negative values of similar magnitudes are obtained, and these are probably a simple consequence of the limited accuracy of the measurement. This implies that the author’s support of thermal diffusion [5] can no longer be maintained. It appears that such also recognised by the author, as the last publication [26] no longer refers to ‘thermal diffusion’: the whole vapour diffusion flow is instead accepted to result from vapour pressure differences only.

Conclusively it has to be inferred here that the analysis by Stephenson [7], using data from [5], is thus similarly disproved. It should moreover be noted that his use of a dry-cup vapour permeability is not entirely representative for measurements that maintain 100 % relative humidity in the entire sample. The vapour permeabilities obtained from Kumaran’s measurements do indeed show that the high relative humidities in the samples significantly increase the vapour permeability.



### **3.2 Galbraith et al. 1998 [9]**

#### **3.2.1 Authors' measurements**

To allow for a wider range in temperature gradients, Galbraith et al. adapted their original cup-based set-up (see above, paragraph 2.1) with a heater mat, heating up the top surface of the samples (via radiation and convection). Measurements were performed on 19.5 mm plasterboard and 25 mm polystyrene insulation, with temperature differences between 5 K and 20 K. An overview can be found in Appendix 1.

#### **3.2.2 Authors' observations**

Measurements at 3 temperature differences allowed the authors to determine a relation between the vapour flow and the temperature gradient (see Figure 6, excluding the 'expected isothermal flows'). They observed "a direct relationship between the measured total moisture flux and the imposed temperature gradient", and deduced that "measurable thermal diffusion does exist for both materials." They derived positive values for  $\delta_T$ , in magnitude about 30 and 55 times bigger than their  $\delta_p$  (for plasterboard and insulation respectively).

#### **3.2.3 Additional analysis**

Figure 6 portrays the measured diffusion flows and the imposed temperature gradients, but confronts those with the expected isothermal flows. It is clear that these isothermal flows totally disturb the earlier link of diffusion flow and temperature gradient. A consistent relation between vapour diffusion and temperature gradient can no longer be detected, and the authors' support for thermal diffusion can therefore not be maintained. Unfortunately, no clear explanation for the observed behaviour can be given.

### **3.3 Peukhuri et al. 2008 [8]**

#### **3.3.1 Authors' measurements**

Peukhuri et al. measured on six materials, five insulation materials (glass fibre, mineral fibre, cellulose, flax, perlite) and cellular concrete. A built-to-purpose set-up, the Megacup [27], was used: the megacup is essentially a small climate chamber, with the large-size sample forming its horizontal top. A standard climate chamber imposed the other set of conditions. The Megacup was usually held at 12-13 °C, while the climate chamber was at 17-21 °C. A number of different vapour pressures were imposed, resulting in 4 to 9 measurement sets for each material. An overview is brought in Appendix 1.

#### **3.3.2 Authors' observations**

A presentation of the measured sets of vapour pressure difference and vapour diffusion flow demonstrated positive non-zero intercepts at 0 Pa vapour pressure difference. Positive should be understood in view of the reference scheme applied in this paper: flows and differences are positive if from the warm to the cold chamber. This led the authors to infer that "there must exist other transport mechanisms than the water vapour pressure-driven one alone" and "the temperature gradient itself is driving the moisture from the warm towards the cold side."

In a further analysis, the measured flows are approximated with a moisture transfer model assuming vapour pressure, temperature and capillary pressure as transport potentials. The measured moisture flows were thus considered combinations of liquid and vapour, the latter driven by gradients in vapour pressure and temperature. This modelling allowed determination of thermal vapour permeabilities  $\delta_T$ : these were primarily positive (except for perlite insulation), and in magnitude about 5 to 50 times bigger than their  $\delta_p$ . It should be noted however that significant liquid transport had to be included to reliably approximate all moisture flow measurements.

### 3.3.3 Additional analysis

This inclusion of liquid transfer is the Achilles heel of the entire analysis. As the measurements took place at RH's from 40 % to 90 %, a liquid flow contribution is plausible: at higher humidities liquid islands may form in the pores and facilitate moisture transfer by 'liquid transfer shortcuts' [28]. When considering vapour diffusion, this is commonly accounted for by a rise in vapour permeability with relative humidity. However, Table 4 in [8] states that the vapour permeabilities applied in the approximation are already wet-cup values. These were measured between 50 % and 93 %, and are consequently representative for the used measurement range. Such wet-cup values already comprise the liquid moisture transfer facilitation. The liquid flows mentioned above were hence added on top of these wet-cup values.

Moreover, the wet-cup values stated in Table 4 in [8] coincide with the dry-cup values, except for the cellular concrete (dry-cup and wet-cup permeabilities are given in Appendix A of [29]). For the five insulation materials, the equality of the dry-cup and wet-cup values implies that the facilitation by liquid islands is insignificant. This does imply that liquid transports can therefore not be called upon for explaining purposes: for those to transpire, the wet-cup permeabilities should have been notably higher than the dry-cup values. Moreover, in such case, dry-cup permeabilities should have been used for the vapour flow component in the approximations. Another explanation is therefore needed to clarify the deviations observed by the authors.

The measurement results on glass fibre and mineral fibre are shown in Figure 7, complemented with the expected isothermal flows. The measured and expected flows obviously disagree and the measured 'vapour flow - vapour pressure difference' relations moreover do not go through the origin. Both observations are attributed to thermal diffusion by the authors. It is also evident however that the deviation between measured and expected flows is not even: the largest deviation is found for the positive pressure differences (high RH in megacup, low RH in room), whereas the deviations at negative pressure differences (intermediate RH in megacup, intermediate RH in room) appear to fade out. With liquid transports ruled out above, it is likely that thermal diffusion would give a constant deviation between the measured and expected flows.

Such observation focuses the attention on the RH-sensors that were used to determine the conditions at both surfaces of the specimen. The 'negative flow' measurements have similar RH levels in the Megacup and in the chamber. A systematic deviation in the RH sensors would affect both sides similarly, with a smaller global effect on the vapour pressure difference. The 'positive flow' measurements on the other hand have high RH in the Megacup and low RH in the chamber. A progressive RH error would strongly affect the vapour pressure difference and consequently the final analysis.

Figures 6.9 and 6.10 in [29] are partially reproduced in Figure 8: they imply that the RH sensors are not very accurate. In two distinct comparisons with dew-point sensors, the deviations were quite large:

- *low RH:* Figure 8 (right) shows that the dew-point sensors yield 49-54% whereas the RH sensors give roughly 49%: a probable underestimation with 2.5% (in absolute RH terms).
- *high RH:* Figure 8 (left) shows that the dew-point sensors give 91% whereas the RH sensors give roughly 84%, Figure 8 (right) shows 87-91% for the dew-point sensors and 72-75% for the RH-sensors: a probable underestimation with 7% and 15% respectively. Here 10% is accepted as representative value.

This evidently represents a progressive error: small at small RH's, larger at larger RH's; such progressive relation is quite common. To correct the reported RH-values, the deviations of 2.5% at 49% RH and of 10% at 74% RH are linearly related:

$$RH_{act} = RH_{mea} + 2.5 + 7.5 \left( \frac{RH_{mea} - 49}{74 - 49} \right) \quad (4)$$

where  $RH_{mea/act}$  is the measured/actual (corrected) RH [%]. Correction of the measured RH's with Eq. (4) transforms Figure 7 to Figure 9. For mineral fibre, the corrected curve now coincides with the expected isothermal curve, hence nullifying all thermal diffusion. For the glass fibre the corrected curve now deviates in the opposite direction, reversing (and decreasing) the 0 Pa intercept. This observation is also valid for the other materials involved: Figure 10 presents the original and corrected measurement results. While the original results show non-zero intercepts and slopes higher than expected from wet-cup permeabilities, the corrected results reveal inconsistent intercepts and slopes in far better agreement with the wet-cup permeabilities.

While the suggested correction, Eq. (4), may not be perfect (bearing in mind the variation already present in Figure 8), it must be concluded that such corrected results are far more plausible than the original results. These corrected results do however no longer support the authors' positive conclusion on the occurrence of thermal diffusion.

### 3.4 Dahl et al., 1996 [6]

To validate two physical models for moisture transport, Dahl et al. [6] made isothermal and non-isothermal moisture transfer experiments on 6.35 and 12.7 mm oriented strand board and 25.4 mm polystyrene insulation. His set-up is similar to Glass [11] and temperature differences ranged from 3 K to 10 K.

The authors set out to validate two moisture flow models, not to verify thermal diffusion. Comparison of numerical and experimental results showed that "isothermal data agree well with the model predictions. There is less agreement with the non-isothermal data, indicating a need for further model development." This statement may be interpreted as support for thermal diffusion. Moreover Glass [11] remarked that Dahl's "results at high RH suggest that temperature is the dominant driving potential". For that reason, the article is treated as 'supporting thermal diffusion'.

The second moisture flow model, by Krus, can though be easily rewritten into a vapour-pressure driven moisture flow equation. The data in Table 5 in [6] indeed indicate that the model gives reasonable predictions for the isothermal cases. Less reliable predictions are obtained for the non-isothermal cases. The deviations between measured and expected flows are highly inconsistent though, also at the high relative humidities mentioned by Glass [11]. The quoted "need for development" should accordingly focus more on the measurements than on the models. In conclusion, no support for thermal diffusion can be detected.

## 4. THERMODYNAMICS OF THERMAL DIFFUSION

After correction, all studies originally supporting thermal diffusion of water vapour in porous materials [5-9] have been brought in line with the studies finding no consistent nor significant evidence for thermal diffusion [9-12]. In most measurements however, the vapour pressure gradients were one to two orders of magnitude larger the temperature gradients, possibly masking the occurrence of thermal diffusion. In the measurements with small vapour pressure gradients, on the other hand, the limited accuracy of the experiments was the restricting factor. To complement the experimental findings on thermal diffusion, the phenomenon is further analysed on thermodynamic grounds and the resulting influence is quantified in an application.

#### 4.1 Thermodynamic elaboration

The earliest prediction and evaluation of thermal diffusion in gasses was based on molecular mechanics [19,20], but a more general description can be obtained by application of irreversible thermodynamics [21]. Irreversible thermodynamics generally applies the second law of thermodynamics, stating that the entropy production in a system undergoing irreversible transformations is positive. The entropy production is commonly written as the bilinear combination of thermodynamic forces and flows [21]:

$$\sigma = -q \frac{\nabla T}{T^2} - j_v \frac{\nabla(\mu_v - \mu_a)_{T,P}}{T} \quad (5)$$

where  $\sigma$  [W/m<sup>3</sup>·K] is the entropy production,  $\mu$  [J/kg] the chemical potential,  $P$  [Pa] the total pressure, and subscripts  $v$  and  $a$  refer to vapour and air respectively.  $\nabla T/T^2$  and  $\nabla(\mu_v - \mu_a)_{T,P}/T$  can consequently be assumed the thermodynamic forces, driving forces for the diffusion of water vapour in air:

$$j_v = -L_{vv} \frac{\nabla(\mu_v - \mu_a)_{T,P}}{T} - L_{vq} \frac{\nabla T}{T^2} \quad (6)$$

where  $L_{vv}$  [kg·K/J·m·s] and  $L_{vq}$  [kg·K/m·s] are phenomenological coefficients.  $L_{vv}$  relates to the standard diffusion, while  $L_{vq}$  represents the thermal diffusion. Essentially, Eq. (6) states that chemical potential and temperature both are transport potentials for the diffusion of water vapour in air.

The chemical potential gradient can be rewritten in terms of mass fractions  $C$  [kg/kg]:

$$C_v = \rho_v / \rho_{a+v} \quad C_a = \rho_a / \rho_{a+v} \quad C_v + C_a = 1 \quad (7)$$

where  $\rho$  [kg/m<sup>3</sup>] is the density. This requires the Gibbs-Duhem relation:

$$C_v \delta \mu_v + C_a \delta \mu_a = 0 \quad (T \text{ and } P \text{ constant}) \quad (8)$$

and the definition for chemical potential of water vapour in air mixtures:

$$\mu_v(T, P) = \mu_v^\circ(T, P) + R_v T \ln(C_v) \quad (9)$$

where  $\mu^\circ$  [J/kg] is a reference value, function of temperature and pressure only, and  $R_v$  [J/kg·K] is the gas constant for water vapour. These transform Eq. (6) to:

$$j_v = -\frac{L_{vv} R_v}{C_v C_a} \nabla C_v - \frac{L_{vq}}{T^2} \nabla T \quad (10)$$

A typical choice for the phenomenological coefficients of Eq. (10) is [21]:

$$j_v = -\rho_{a+v} D \nabla C_v + \rho_{a+v} D' C_v C_a \nabla T \quad (11)$$

where  $D$  [m<sup>2</sup>/s] is the standard water vapour-air diffusivity and  $D'$  [m<sup>2</sup>/K·s] is the diffusivity for temperature gradients. These particular choices reflect the respective independence and dependence of the diffusivities on the vapour mass fraction [21] and the 'negative' thermal diffusion experienced by the lighter vapour molecules [22]. Generally in thermal diffusion the heavy constituent diffuses with the temperature gradient, whereas the light constituent diffuses against the temperature gradient. For air, the dominant N<sub>2</sub> molecules form the heavy constituent, while H<sub>2</sub>O is the light constituent. The thermal diffusivity  $D'$  is usually related to the standard diffusivity  $D$  via the thermal diffusion factor  $\alpha$  [-] [21]:

$$j_v = -\rho_{a+v} D \nabla C_v + \rho_{a+v} \frac{\alpha}{T} D C_v C_a \nabla T \quad (12)$$

For building science applications, the global pressure is commonly presumed constant, and equal to the atmospheric pressure. This finally allows stating:

$$j_v = -\frac{D}{R_v T} \nabla p_v + \rho_{a+v} \frac{\alpha}{T} D C_v C_a \nabla T = j_p + j_T = -\delta_p \nabla p_v - \delta_T \nabla T \quad (13)$$

Diffusion of water vapour in air may thus result from vapour pressure gradients as well as from temperature gradients, hence confirming Eq. (1).

The relative importance of the standard and thermal water vapour diffusion can be estimated from the ratio of their transport coefficients:

$$\frac{\delta_T}{\delta_p} = \left( -\rho_{a+v} \frac{\alpha}{T} D C_v C_a \right) / \left( \frac{D}{R_v T} \right) = -\rho_{a+v} \alpha C_v C_a R_v \quad (14)$$

Unfortunately, no measured  $\alpha$  value is available for water vapour and air mixtures, and it thus needs to be estimated. Based on the molecular model of [19], one gets values from 0.13 to 0.16, comparing well with other binary gas mixtures [22]: 0.15 is assumed as typical value here. For vapour pressures from 500 to 3000 Pa – representative for many building science applications – Eq. (10) results in ratio's from -0.25 to -1.5. The thermal permeability  $\delta_T$  is thus of the same order of magnitude as  $\delta_p$ , but negative, indicating thermal diffusion transpiring from low to high temperatures. [23] corroborates that the thermal diffusion factor  $\alpha$  remains valid when considering porous materials, so such ratio's are equally valid for vapour diffusion in porous materials.

## 4.2 Application example

To exemplify the potential effects of thermal diffusion, vapour diffusion in a simple construction with exterior insulation is quantified. The construction consists of: 20 cm concrete (2.60 W/m·K & 2.0 10<sup>-12</sup> kg/m·s·Pa), 10 cm mineral wool insulation (0.04 W/m·K & 1.5 10<sup>-10</sup> kg/m·s·Pa), 1 cm stucco (0.30 W/m·K & 3.0 10<sup>-11</sup> kg/m·s·Pa) (interior to exterior; values between brackets are thermal conductivity and standard permeability  $\delta_p$ ). Interior and exterior surface resistances are 0.13 m<sup>2</sup>·K/W & 5.0 10<sup>7</sup> m<sup>2</sup>·s·Pa/kg and 0.04 m<sup>2</sup>·K/W & 6.7 10<sup>6</sup> m<sup>2</sup>·s·Pa/kg. Interior and exterior conditions are respectively 20 °C & 1200 Pa and -10 °C & 200 Pa. All thermal permeabilities are taken equal in magnitude to the standard permeabilities but negative, for ease of calculation.

First, the vapour pressure profile and the vapour diffusion flow are quantified based on standard diffusion only, hence solely driven by vapour pressure gradients. Results can be found in Table 1. While for the concrete and stucco layers the vapour pressure gradient is far larger than the temperature gradient, this is not valid for the insulation layer: the temperature gradient is 270 K/m, while the gradient in vapour pressure is 66 Pa/m. With standard and thermal vapour permeabilities of equal magnitude, thermal diffusion may have a serious impact. The profile and flow are thus recalculated with inclusion of thermal diffusion, the results of which can be found in Table 1 as well. Thermal diffusion raises the vapour pressure gradient over the insulation to 334 Pa/m, larger than the temperature gradient. This is of course necessary to ensure a continuous flow from interior to exterior. The influence of thermal diffusion should hence not be assessed from a single layer; instead the complete construction is to be considered.

The more fascinating outcome is the influence of thermal diffusion on the global diffusion flow: including negative thermal permeabilities of equal magnitude lessens the diffusion flow with (only) 3 %. This -3% equals the ratio of the 30 K temperature difference and the 1000 Pa vapour pressure difference over the wall construction, multiplied with the ratio of the thermal permeability to the standard permeability, Eq. (14). It can easily be shown that this conclusion remains valid for other conditions and other constructions.

## 5. DISCUSSION

### 5.1 Vapour pressure

Most research on thermal diffusion in porous materials [5-12] is experimental, and all investigations apply the same measurement concept. The diffusion flow through a single layer of material under combined gradients of vapour pressure and temperature is measured, and compared to the respective isothermal diffusion flow. Multiple studies [5-9] reanalysed here originally supported thermal diffusion, by detecting consistent and significant deviations from isothermal diffusion. All of them were however shown to be flawed. The positive '0 Pa' intercept, in [5], was shown to not be generally valid. The perceived relation between vapour diffusion and temperature gradients in [9] was virtually annulled by introducing the expected isothermal flows. All deviation between measured and expected flows in [8] vanished after correcting the measurement RH's. Differences between measured and isothermal flows in [6] were finally shown to be inconsistent. Reinterpretation of their results brought them in line with the other studies [9-12], that already corroborated that no consistent nor significant thermal diffusion is to be detected. The examination here has hence succeeded in reconciling their previously contradictory findings, to reach a convergent conclusion: no consistent nor significant thermal diffusion can be detected.

This in the end does not come as a surprise. Whereas the thermodynamic elaboration points to vapour pressure and temperature as possible transport potentials, the phenomenological coefficients imply that only minor negative thermal diffusion occurs. In the measurements analysed above, temperature differences were generally one to two orders of magnitude lower than the vapour pressure differences. With the standard and thermal vapour permeabilities of comparable magnitude, the effect of thermal diffusion is minimal, remaining undetectable with the achievable measurement accuracy. In the measurements with minimal vapour pressure differences, on the other hand, the small flow similarly remains undetectable.

This conclusion is more generally valid for vapour diffusion in building science applications: often vapour pressure differences over a construction are far larger than temperature differences, with resultantly a negligible role for thermal diffusion. It was shown above that the effect of thermal diffusion is proportional to the ratio of the temperature difference to the vapour pressure difference, which is mostly quite small. In the exceptional case of a dominant temperature difference the resultant flow would be negligible as well. It can hence cautiously be concluded that thermal diffusion of water vapour in porous materials can be considered insignificant, and that vapour pressure is the sole significant transport potential for diffusion of water vapour in porous materials:

$$j_v = j_p = -\delta_p \nabla p_v \quad (15)$$

for both isothermal and non-isothermal conditions. Only a single transport parameter is hence to be determined, obtainable by simple measurement of vapour flow and vapour pressure difference.

### 5.2 Other potentials

Under isothermal conditions this vapour pressure gradient can easily be translated to a vapour concentration gradient, in line with the original law of Fick [30], derived from isothermal experiments as well. Several authors do though equally use this concentration gradient for non-isothermal conditions [31,32], and then imply that a 'thermal diffusion' term is required. This term does not represent diffusion resulting from temperature gradients though. It is a mere parasitic diffusion term, resulting from choosing a transport potential differing from the true potential:

$$j_v = -\delta_p \nabla p_v = -\delta_p R_v T \nabla \rho_v - \delta_p R_v \rho_v \nabla T = -D_p \nabla \rho_v - \varepsilon D_p \nabla T \quad (16)$$

where  $\epsilon$  [ $\text{s/m}^2 \cdot \text{K}$ ] is an alternative thermal diffusion factor. The same is true when relative humidity or moisture content is used as transport potential. The prior thermodynamic elaboration shows that chemical potential, mass fraction and vapour pressure are equivalent: their further translation to vapour concentration is only valid though for isothermal conditions.

For example, Qin et al. [31] use the second expression in Eq. (16) for their vapour diffusion measurements on Gotland sandstone: they report  $1.168 \cdot 10^{-6} \text{ m}^2/\text{s}$  and  $3.16 \cdot 10^{-4} / \text{K}$  as resulting  $D_p$  and  $\epsilon$  values. The determination of the thermal transport parameter  $\epsilon$  is quite challenging: both an isothermal and a non-isothermal measurement are required, for which vapour flows, temperature profiles and vapour concentrations profile are to be quantified. Temperature profiles require installing thermocouples in the sample, whereas the vapour concentration profiles can only be obtained indirectly, from relative humidity or moisture content profiles. Those in turn necessitate the slicing of the sample prior to or after the test. The determination of  $D_p$  and  $\epsilon$  is hence far more demanding than the determination of  $\delta_p$ .

The non-isothermal measurement applies  $0.532 \text{ kg/m}^4$  and  $259 \text{ K/m}$  as  $\nabla p_v$  and  $\nabla T$ ; the isothermal measurement applies  $0.130 \text{ kg/m}^4$  as  $\nabla p_v$  (see Figures 4, 9, 10 in [31]). The non-isothermal experiment hence yields  $6.21 \cdot 10^{-7}$  and  $9.56 \cdot 10^{-8} \text{ kg/m}^2 \cdot \text{s}$  as  $\nabla p_v$  and  $\nabla T$  vapour flows respectively. The alleged thermal diffusion appears to play a serious role, responsible for 13 % of the total flow. Although, the non-isothermal measurement can also be characterised with a  $7.76 \cdot 10^4 \text{ Pa/m}$   $\nabla p_v$ . This finally yields  $9.2 \cdot 10^{-12} \text{ kg/m} \cdot \text{s} \cdot \text{Pa}$  as standard permeability  $\delta_p$ . The isothermal measurement, on the other hand, leads to  $8.5 \cdot 10^{-12} \text{ kg/m} \cdot \text{s} \cdot \text{Pa}$ .

With relative humidities between 65% and 85% (isothermal) and between 65% and 95% (non-isothermal), the measurements do not relate to pure vapour diffusion solely: liquid transfer enhancement, via the liquid islands in the small pores, must play a role as well. This influence is more important at higher humidities, undoubtedly explaining the slightly higher  $\delta_p$  value for the non-isothermal measurement. Ultimately, both  $\delta_p$  values can be assumed consistent.

The conclusions on the insignificance of thermal diffusion might not apply, as this case deviates from pure vapour diffusion. The resulting isothermal and non-isothermal  $\delta_p$  do however confirm the validity of Eq. (15), and thus the absence of actual thermal diffusion. The alleged importance of the thermal transport must therefore be assumed a mere consequence of deviating from the true transport potential. To conclude, description of (non-)isothermal diffusion with vapour concentration and temperature gradients, and the associated method for transport parameter determination, is an undesirable complication: a simple description with vapour pressure, with an easily obtained transport parameter, does actually suffice. Similar conclusions can be drawn for the results shown in [32].

## 6. CONCLUSIONS

Reliable evaluation of moisture transport in porous materials requires a correct description of moisture flow phenomena and their transport potentials. In that respect, thermal diffusion of water vapour has been strongly disputed: several studies [9-12] corroborate that vapour pressure is the sole significant transport potential, while multiple other studies [5-9] infer that temperature gradients may also yield consistent and significant diffusion of water vapour.

This last statement has been clearly disproved in this paper. The latter studies were all shown to be flawed, and a (corrected) reinterpretation brought them in line with the former studies: no consistent nor significant thermal diffusion can be detected, at least not with the used measurement principles and conditions. The thermodynamic elaboration complemented and generalised that conclusion. It indicated that negative thermal diffu-

sion is to be expected, although with a magnitude that is negligible for most building science applications. Perhaps the time has come to finally leave 'wishful thinking' behind, and refer 'significant thermal diffusion of water vapour in porous materials' to the realm of fiction.

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## APPENDIX 1

### Measurement results 2.1

	particle board				polystyrene insulation		
T cup [°C]	15.0	15.0	15.0	15.0	15.0	15.0	15.0
RH cup [%]	76	43	36	3	76	43	3
$p_v$ cup [Pa]	1298	735	615	51	1298	735	51
T chamber [°C]	23.0	23.0	23.0	23.0	23.0	23.0	23.0
RH chamber [%]	60	60	60	60	60	60	60
$p_v$ chamber [Pa]	1689	1689	1689	1689	1689	1689	1689
$p_v$ difference [Pa]	390	954	1074	1638	390	954	1638
measured flow [ $10^{-8}$ kg/m <sup>2</sup> s]	12.9	29.9	31.7	48.6	3.5	7	13.3
stand. deviation [ $10^{-8}$ kg/m <sup>2</sup> s]	0.4	2.2	3	3.7	1	1.2	3.4
isothermal flow [ $10^{-8}$ kg/m <sup>2</sup> s]	11.7	28.6	32.2	49.1	3.56	8.7	14.9
thermal diffusion [ $10^{-8}$ kg/m <sup>2</sup> s]	1.19	1.27	-0.51	-0.53	-0.06	-1.7	-1.63

Values in regular font are taken from the original article; values in italic have been calculated in this paper.

All calculated flows use the vapour pressure difference between cup and chamber, but neglect the surface resistances: for materials with the given permeability and thickness, these are negligible. The permeabilities are taken constant based on values in [9]: respectively  $3.60 \cdot 10^{-12}$  and  $2.28 \cdot 10^{-12}$  kg/msPa for the particle board and polystyrene insulation. The authors instead apply a fitted RH-dependent  $\delta_p$  for the particle board; deviations between the two approaches are minimal though.

To exemplify the global standard deviation on the isothermal flows, find the required results underneath. The standard deviation on each of the permeabilities is converted into a standard deviation on the flow. While there is a weak relation with the flow magnitude, these are still aggregated into one value:  $1.9 \cdot 10^{-8}$  and  $1.5 \cdot 10^{-8}$  kg/m<sup>2</sup>s for the particle board and polystyrene insulation respectively.

	particle board				polystyrene insulation			
RH cup [%]	3	79	93	100	3	79	93	100
$p_v$ cup [Pa]	84	2224	2618	2815	84	2224	2618	2815
RH chamber [%]	60	60	60	60	50	60	60	60
$p_v$ chamber [Pa]	1689	1689	1689	1689	1407	1689	1689	1689
$p_v$ difference [Pa]	-1604	535	929	1126	-1323	535	929	1126
measured $\delta_p$ [ $10^{-12}$ kg/msPa]	3.66	3.89	5.59	6.43	2.28	2.11	2.50	2.12
stand. deviation [ $10^{-12}$ kg/msPa]	0.10	0.27	0.22	0.32	0.42	0.31	0.39	0.29
isothermal flow [ $10^{-8}$ kg/m <sup>2</sup> s]	-48.93	17.34	43.27	60.33	-12.06	4.51	9.29	9.55
stand. deviation [ $10^{-8}$ kg/m <sup>2</sup> s]	1.34	1.20	1.70	3.00	2.22	0.66	1.45	1.31

Values in regular font are taken from the original article; values in italic have been calculated in this paper.

### Measurement results 2.2

	oriented strand board									
T bottom [°C]	35.0	24.8	25.1	10.0	35.1	30.0	35.0	35.0	20.1	30.0
RH bottom [%]	22.8	45	45.1	32	25.2	34	26.1	31.9	44.9	52.4
$p_v$ bottom [Pa]	1285	1413	1436	394	1427	1446	1469	1795	1057	2234
T top [°C]	25.0	35.0	35.1	5.1	25.0	25.0	25.0	25.1	25.0	25.0
RH top [%]	44.9	22.8	25.2	45.1	45.0	45.0	45.0	45.0	45.0	45.0

$p_v$ top [Pa]	1429	1284	1427	395	1427	1429	1426	1434	1429	1432
$p_v$ difference [Pa]	-144	-129	-9	-1	1	17	43	360	372	802
measured flow [ $10^{-8}$ kg/m <sup>2</sup> s]	-0.77	-1.95	0.48	0.33	0.55	0.62	0.79	2.97	3.04	6.74
standard deviation [ $10^{-8}$ kg/m <sup>2</sup> s]	0.06	0.07	0.02	0.07	0.05	0.07	0.26	0.63	0.34	0.47
isothermal flow [ $10^{-8}$ kg/m <sup>2</sup> s]	-1.27	-1.14	-0.08	-0.01	0.01	0.15	0.38	3.19	3.29	7.11
thermal diffusion [ $10^{-8}$ kg/m <sup>2</sup> s]	0.5	-0.81	0.56	0.34	0.54	0.47	0.41	-0.22	-0.25	-0.37

Values in regular font are taken from the original article; values in italic have been calculated in this paper.

All calculated flows use the vapour pressure difference between cup and chamber, but neglect the surface resistances: for materials with the given permeability and thickness, these are negligible. The given standard deviations differ from the 'uncertainties' given by the authors. The values here were calculated from the measured flow values given in Appendix B in [10]. It has to be noted that two corrections to the reported data have been made based on Appendix B: in test 8 the bottom temperature is 35.0 °C (instead of 30 °C), while in test 2 the top and bottom conditions are swapped.

The isothermal flows are estimated with a permeability derived from the two isothermal tests (Table 1 in [10]), averaging to  $5.63 \cdot 10^{-13}$  kg/msPa. The standard deviation on both isothermal experiments aggregates to  $1.9 \cdot 10^{-9}$  kg/m<sup>2</sup>s.

### Measurement results 2.3

	chipboard			con- crete	MDF			mor- tar	phenolic foam		
average RH [%]	42	59	76	59	46	61	73	61	45	57	57
average T [°C]	27	27	25	27	26	27	26	27	25	20	26
T difference [°C]	9	8	9	7	8	9	10	7	11	6	10
measurement $\delta_p$ [ $10^{-12}$ kg/msPa]	4.2	3.9	6.2	4.6	8.6	9.7	12	5.7	3	2.8	3.2
cup measured $\delta_p$ [ $10^{-12}$ kg/msPa]	4.6	4.8	5.6	6.5	7.2	7.5	8	8.9	2.5	2.6	2.7
relative difference [-]	-9%	-19%	11%	-29%	19%	29%	50%	-36%	20%	8%	19%
	plasterboard			plywood			skinned XPS	unskinned XPS			
average RH [%]	42	56	75	48	54	75	47	55	46	56	
average T [°C]	25	25	26	26	28	26	27	28	27	28	
T difference [°C]	10	10	9	8	8	9	10	9	9	8	
measurement $\delta_p$ [ $10^{-12}$ kg/msPa]	34	35	41	1.4	2.3	3.5	2.1	1.3	1.4	1.9	
cup measured $\delta_p$ [ $10^{-12}$ kg/msPa]	24	26	32	1.5	1.6	2.6	2.5	2.1	2	2.6	
relative difference [-]	42%	35%	28%	-7%	44%	35%	-16%	-38%	-30%	-27%	

Values in regular font are taken from the original article; values in italic have been calculated in this paper.

### Measurement results 3.1

	d = 0.0855 m, $\rho = 17$ kg/m <sup>3</sup>				d = 0.0547 m, $\rho = 30$ kg/m <sup>3</sup>					
$T_+$ [°C]	39.4	39.5	50.7	56.1	34.7	34.8	39.8	44.2	48.7	53.1
$T_-$ [°C]	11.6	-1.5	12.1	16.9	13.8	13.8	14.0	14.2	14.4	14.6
$p_{v,+}$ [Pa]	7131	7169	12738	16542	5522	5553	7285	9177	11534	14324
$p_{v,-}$ [Pa]	1364	545	1409	1929	1576	1576	1596	1617	1638	1659
$q_{wet}$ [W/m <sup>2</sup> ]	38.9	48.0	67.5	81.2	39.6	39.6	53.8	67.9	84.2	102.8
$q_{dry}$ [W/m <sup>2</sup> ]	12.6	18.1	18.1	18.9	13.9	14.0	17.4	20.6	23.7	27.0
$j_v$ [ $10^{-6}$ kg/m <sup>2</sup> s]	10.67	12.01	20.09	25.46	10.44	10.40	14.80	19.25	24.64	30.90

	d = 0.025 m, $\rho = 45 \text{ kg/m}^3$					d = 0.050 m, $\rho = 45 \text{ kg/m}^3$				
$T_+ [^\circ\text{C}]$	34.3	37.0	41.5	47.1	51.5	33.3	37.8	42.3	48.2	52.9
$T_- [^\circ\text{C}]$	14.2	14.4	14.8	15.3	15.6	13.2	13.3	13.5	13.8	14.2
$p_{v,+} [\text{Pa}]$	5422	6279	8001	10664	13269	5134	6557	8348	11271	14193
$p_{v,-} [\text{Pa}]$	1621	1643	1685	1738	1775	1519	1533	1553	1579	1619
$q_{\text{wet}} [\text{W/m}^2]$	70.0	81.7	102.5	131.4	158.4	38.1	49.9	63.9	84.8	103.7
$q_{\text{dry}} [\text{W/m}^2]$	28.0	31.5	37.6	45.4	51.6	13.6	16.7	19.9	24.0	27.3
$j_v [10^{-6} \text{ kg/m}^2\text{s}]$	17.05	20.40	26.43	35.05	43.54	9.92	13.48	17.90	24.75	31.13
	d = 0.075 m, $\rho = 45 \text{ kg/m}^3$					d = 0.100 m, $\rho = 45 \text{ kg/m}^3$				
$T_+ [^\circ\text{C}]$	33.5	38.0	42.8	48.6	53.4	33.4	38.0	43.1	49.0	53.6
$T_- [^\circ\text{C}]$	12.9	13.0	13.2	13.4	13.5	12.7	12.8	13.1	13.1	13.2
$p_{v,+} [\text{Pa}]$	5186	6618	8565	11499	14555	5146	6629	8677	11743	14689
$p_{v,-} [\text{Pa}]$	1487	1502	1519	1537	1551	1475	1484	1507	1512	1524
$q_{\text{wet}} [\text{W/m}^2]$	27.2	35.9	47.1	62.7	78.7	21.0	28.1	37.4	50.8	62.9
$q_{\text{dry}} [\text{W/m}^2]$	9.3	11.3	13.6	16.3	18.9	7.0	8.6	10.4	12.5	14.2
$j_v [10^{-6} \text{ kg/m}^2\text{s}]$	7.27	10.00	13.62	18.90	24.38	5.67	7.93	10.99	15.57	19.83
	d = 0.125 m, $\rho = 45 \text{ kg/m}^3$					d = 0.055 m, $\rho = 30 \text{ kg/m}^3$				
$T_+ [^\circ\text{C}]$	33.7	38.3	43.2	49.2	53.7	34.7	39.8	44.2	48.7	53.1
$T_- [^\circ\text{C}]$	12.8	12.8	13.0	13.2	13.0	13.8	13.9	14.2	14.4	14.6
$p_{v,+} [\text{Pa}]$	5224	6733	8722	11825	14746	5544	7285	9191	11546	14352
$p_{v,-} [\text{Pa}]$	1480	1480	1496	1519	1503	1576	1595	1617	1639	1659
$q_{\text{wet}} [\text{W/m}^2]$	17.4	23.7	30.7	42.2	52.8	39.6	53.8	67.9	84.2	102.8
$q_{\text{dry}} [\text{W/m}^2]$	5.7	7.0	8.4	10.1	11.8	13.9	17.4	20.6	23.7	27.0
$j_v [10^{-6} \text{ kg/m}^2\text{s}]$	4.76	6.80	9.10	13.08	16.67	8.26	11.71	15.23	19.49	24.44
	d = 0.051 m, $\rho = 66 \text{ kg/m}^3$					d = 0.055 m, $\rho = 117 \text{ kg/m}^3$				
$T_+ [^\circ\text{C}]$	30.7	34.6	37.5	40.2	42.8	45.3	36.1	39.1	41.9	44.7
$T_- [^\circ\text{C}]$	13.6	13.7	13.9	14.0	14.1	14.2	12.2	12.2	12.3	12.5
$p_{v,+} [\text{Pa}]$	4411	5510	6438	7458	8565	9751	5968	7036	8154	9455
$p_{v,-} [\text{Pa}]$	1555	1569	1586	1605	1612	1620	1421	1427	1437	1450
$q_{\text{wet}} [\text{W/m}^2]$	30.7	39.6	47.6	55.4	63.4	72.7	41.8	49.9	57.4	67.4
$q_{\text{dry}} [\text{W/m}^2]$	11.1	13.6	15.4	17.3	18.9	21.0	15.4	17.4	19.1	21.1
$j_v [10^{-6} \text{ kg/m}^2\text{s}]$	6.30	8.36	10.36	12.26	14.32	16.65	8.48	10.44	12.31	14.89
	continued		d = 0.154 m, $\rho = 45 \text{ kg/m}^3$							
$T_+ [^\circ\text{C}]$	47.5	50.3	36.4	40.9	45.6	50.0	53.4			
$T_- [^\circ\text{C}]$	12.6	12.7	11.7	11.8	11.8	11.9	11.9			
$p_{v,+} [\text{Pa}]$	10859	12501	6074	7723	9866	12360	14576			
$p_{v,-} [\text{Pa}]$	1459	1472	1376	1386	1388	1391	1396			
$q_{\text{wet}} [\text{W/m}^2]$	76.8	87.3	17.9	23.3	29.9	37.2	43.9			
$q_{\text{dry}} [\text{W/m}^2]$	22.8	24.6	5.9	7.1	8.6	9.6	10.4			
$j_v [10^{-6} \text{ kg/m}^2\text{s}]$	17.38	20.18	3.85	5.21	6.85	8.88	10.78			
	d = 0.025 m, $\rho = 45 \text{ kg/m}^3$					d = 0.051 m, $\rho = 45 \text{ kg/m}^3$				
$T_+ [^\circ\text{C}]$	31.9	34.2	37.9	42.1	45.6	32.0	35.7	39.9	44.8	48.6
$T_- [^\circ\text{C}]$	16.8	17.7	19.0	20.8	22.5	14.8	15.5	16.4	17.8	19.1
$p_{v,+} [\text{Pa}]$	4730	5389	6600	8261	9866	4749	5848	7355	9489	11522
$p_{v,-} [\text{Pa}]$	1920	2027	2203	2454	2734	1683	1768	1869	2037	2210
$q_{\text{wet},+} [\text{W/m}^2]$	69.5	81.1	101.6	130.5	156.6	37.7	49.4	63.0	83.5	101.5

$q_{\text{wet},-}$ [W/m <sup>2</sup> ]	70.5	82.5	103.7	133.3	160.1	38.4	50.4	64.7	86.2	105.7
$q_{\text{dry}}$ [W/m <sup>2</sup> ]	20.9	23.1	26.6	30.5	33.2	11.6	14.0	16.2	18.9	20.9
$j_v$ [10 <sup>-6</sup> kg/m <sup>2</sup> s]	19.98	23.94	31.05	41.48	51.31	10.74	14.62	19.41	26.93	33.81
d = 0.077 m, $\rho = 45$ kg/m <sup>3</sup>						d = 0.102 m, $\rho = 45$ kg/m <sup>3</sup>				
$T_+$ [°C]	32.4	36.6	41.0	45.9	50.0	32.6	37.0	41.5	46.8	50.8
$T_-$ [°C]	14.0	14.6	15.3	16.3	17.4	13.7	14.2	14.7	15.7	16.4
$p_{v,+}$ [Pa]	4882	6140	7772	10044	12354	4917	6265	7976	10530	12858
$p_{v,-}$ [Pa]	1600	1669	1745	1857	1990	1566	1618	1675	1781	1870
$q_{\text{wet},+}$ [W/m <sup>2</sup> ]	27.0	35.5	46.5	61.5	77.0	20.6	27.8	36.8	49.8	61.3
$q_{\text{wet},-}$ [W/m <sup>2</sup> ]	27.6	36.4	47.7	63.7	80.2	21.6	28.5	37.8	51.7	64.3
$q_{\text{dry}}$ [W/m <sup>2</sup> ]	8.3	10.0	11.8	13.7	15.5	6.4	7.8	9.3	10.9	12.1
$j_v$ [10 <sup>-6</sup> kg/m <sup>2</sup> s]	7.71	10.57	14.39	19.94	25.79	5.97	8.28	11.41	16.25	20.69
d = 0.128 m, $\rho = 45$ kg/m <sup>3</sup>						d = 0.154 m, $\rho = 45$ kg/m <sup>3</sup>				
$T_+$ [°C]	32.9	37.3	41.9	47.4	51.4	32.9	37.6	42.0	47.6	51.7
$T_-$ [°C]	13.4	13.8	14.3	15.3	15.9	13.2	13.7	14.1	14.6	15.1
$p_{v,+}$ [Pa]	5004	6372	8150	10838	13217	5001	6490	8192	10936	13380
$p_{v,-}$ [Pa]	1544	1581	1635	1736	1804	1522	1567	1607	1665	1720
$q_{\text{wet},+}$ [W/m <sup>2</sup> ]	17.2	23.5	30.4	41.6	51.8	14.0	19.4	25.2	35.6	45.6
$q_{\text{wet},-}$ [W/m <sup>2</sup> ]	17.5	23.7	30.9	43.3	53.3	15.1	20.5	27.3	37.0	45.3
$q_{\text{dry}}$ [W/m <sup>2</sup> ]	5.3	6.4	7.6	9.0	10.3	4.5	5.5	6.5	7.7	8.6
$j_v$ [10 <sup>-6</sup> kg/m <sup>2</sup> s]	4.90	6.99	9.37	13.64	17.22	4.10	5.87	8.05	11.67	15.03

Values in regular font are taken from the original article; values in italic have been calculated in this paper.

The transformation from heat flows to vapour flows follows Eq. (12) or (13), involving the vaporisation heat of water and the specific heat of water vapour:

$$h_{w \rightarrow v} = 2.501 \cdot 10^6 - 2.373 \cdot 10^3 T \quad [\text{J} / \text{kg}] \quad (17)$$

$$c_v = 1.812 \cdot 10^3 \quad [\text{J} / \text{kgK}]$$

### Measurement results 3.2

	plasterboard			polystyrene insulation		
temperature difference [K]	5.0	7.2	10.1	8.8	13.6	19.7
thermal gradient [K/m]	256	366	516	350	546	786
measured flow [10 <sup>-8</sup> kg/m <sup>2</sup> s]	152	161	176	6	8	10
standard deviation [10 <sup>-8</sup> kg/m <sup>2</sup> s]	5.7	4.8	7.1	0.610	0.480	0.810
isothermal flow [10 <sup>-8</sup> kg/m <sup>2</sup> s]	169	169	169	7.38	7.38	7.38
thermal diffusion [10 <sup>-8</sup> kg/m <sup>2</sup> s]	-16.9	-7.9	7.1	-1.64	0.44	2.37

Values in regular font are taken from the original article; values in italic have been calculated in this paper.

Isothermal flows were calculated from the constant vapour pressure difference maintained in the test (1098 Pa) and the vapour permeabilities of the materials ( $3.07 \cdot 10^{-11}$  and  $1.68 \cdot 10^{-12}$  kg/msPa for plasterboard and polystyrene insulation respectively). The given vapour pressure difference is surface-to-surface, so no surface resistances need to be accounted for. The global standard deviations on the isothermal flows were derived as above from data given in Table 8 in [9], and aggregated to  $7.1 \cdot 10^{-8}$  and  $9.9 \cdot 10^{-9}$  kg/m<sup>2</sup>s.

### Measurement results 3.3

	glass fibre, d = 0.069 m					cell. concrete, d = 0.093 m			
T Megacup [°C]	12.3	12.2	12.1	12.0	12.1	12.9	12.9	13.0	12.8
RH Megacup [%]	64.0	70.1	79.5	76.8	75.9	70.4	78.4	82.6	85.6
$p_v$ Megacup [Pa]	917	998	1124	1079	1073	1049	1169	1239	1267
T chamber [°C]	18.2	18.2	18.1	17.7	17.7	20.9	20.9	20.9	20.8
RH chamber [%]	55.0	56.6	55.6	45.6	43.5	54.0	54.3	54.8	45.6
$p_v$ chamber [Pa]	1152	1185	1157	925	883	1337	1345	1357	1122
$p_v$ difference [Pa]	-235	-187	-33	154	191	-288	-176	-118	145
RH Megacup [%] corrected	71.0	78.9	91.2	87.6	86.5	79.3	89.7	95.2	99.1
RH chamber [%] corrected	59.3	61.4	60.1	47.1	44.4	59.0	59.5	60.2	46.4
$p_v$ Megacup [Pa] corrected	1017	1124	1289	1231	1223	1182	1337	1428	1467
$p_v$ chamber [Pa] corrected	1242	1285	1250	955	900	1461	1472	1491	1142
$p_v$ difference [Pa] corrected	-224	-162	39	276	323	-279	-135	-63	325
measured flow [ $10^{-8}$ kg/m <sup>2</sup> s]	-55.28	-39.84	9.56	67.97	79.55	-7.23	-3.50	-1.63	8.44
isothermal flow [ $10^{-8}$ kg/m <sup>2</sup> s]	-63.30	-51.30	-0.40	59.50	67.80	-14.20	-4.42	-1.06	30.40
	mineral fibre, d = 0.100 m								
T Megacup [°C]	12.5	12.4	12.4	12.2	12.1	12.2	12.2	12.1	12.2
RH Megacup [%]	61.7	63.1	64.9	69.8	79.3	79.0	78.6	75.5	79.5
$p_v$ Megacup [Pa]	896	910	936	994	1121	1125	1119	1068	1132
T chamber [°C]	20.8	20.8	20.8	20.8	20.9	20.9	20.6	20.4	20.2
RH chamber [%]	51.8	52.0	52.2	52.4	49.1	49.1	41.6	38.4	38.6
$p_v$ chamber [Pa]	1275	1280	1285	1290	1216	1216	1011	922	916
$p_v$ difference [Pa]	-379	-370	-349	-296	-95	-91	108	146	216
RH Megacup [%] corrected	68.0	69.8	72.2	78.5	90.9	90.5	90.0	86.0	91.2
RH chamber [%] corrected	55.1	55.4	55.7	55.9	51.6	51.6	41.9	37.7	38.0
$p_v$ Megacup [Pa] corrected	987	1007	1041	1118	1285	1288	1281	1215	1297
$p_v$ chamber [Pa] corrected	1357	1363	1370	1376	1279	1279	1018	906	901
$p_v$ difference [Pa] corrected	-370	-356	-329	-258	7	10	263	310	397
measured flow [ $10^{-8}$ kg/m <sup>2</sup> s]	-67.65	-65.21	-60.20	-47.27	1.23	1.77	48.07	56.67	72.59
isothermal flow [ $10^{-8}$ kg/m <sup>2</sup> s]	-79.20	-70.00	-66.90	-50.60	2.39	0.11	51.20	57.50	85.90
	cellulose insulation, d = 0.100 m					flax insulation, d = 0.090 m			
T Megacup [°C]	12.5	12.7	12.3	12.2	12.5	12.6	12.2	15.2	12.3
RH Megacup [%]	59.5	70.9	77.7	80.0	68.0	67.5	82.3	57.8	88.8
$p_v$ Megacup [Pa]	864	1043	1113	1139	987	986	1172	1000	1272
T chamber [°C]	20.7	20.7	20.7	20.8	21.0	21.0	21.1	20.9	21.1
RH chamber [%]	52.5	52.8	48.9	45.6	53.4	51.9	53.3	45.9	54.0
$p_v$ chamber [Pa]	1284	1292	1196	1122	1331	1293	1336	1137	1354
$p_v$ difference [Pa]	-420	-249	-83	16	-343	-307	-165	-136	-81
RH Megacup [%] corrected	67.3	84.4	94.6	98.0	76.2	75.6	94.8	62.9	103.2
RH chamber [%] corrected	56.8	57.2	51.4	46.4	58.1	55.9	58.0	46.9	59.0
$p_v$ Megacup [Pa] corrected	976	1241	1355	1395	1106	1104	1349	1089	1479
$p_v$ chamber [Pa] corrected	1388	1399	1256	1142	1448	1392	1453	1160	1479
$p_v$ difference [Pa] corrected	-412	-158	99	253	-341	-287	-103	-71	0
measured flow [ $10^{-8}$ kg/m <sup>2</sup> s]	-	-17.42	10.86	27.83	-56.89	-47.91	-17.25	-11.84	0.03
isothermal flow [ $10^{-8}$ kg/m <sup>2</sup> s]	-	-30.30	-0.15	19.30	-52.90	-48.80	-14.90	-15.20	-0.67

	perlite insulation, d = 0.123 m				perlite insulation, d = 0.083 m			
T Megacup [°C]	13.0	13.0	13.0	13.0	13.0	12.9	13.0	12.9
RH Megacup [%]	52.7	69.1	71.4	73.5	69.8	80.5	74.4	68.9
$p_v$ Megacup [Pa]	791	1037	1071	1103	1047	1200	1116	1027
T chamber [°C]	20.9	20.9	21.1	20.8	21.1	21.1	20.7	21.1
RH chamber [%]	52.6	52.8	46.2	45.0	52.2	51.5	49.1	44.8
$p_v$ chamber [Pa]	1303	1308	1158	1108	1309	1291	1201	1123
$p_v$ difference [Pa]	-512	-271	-87	-5	-261	-91	-85	-96
RH Megacup [%] corrected	56.3	77.6	80.6	83.4	78.5	92.5	84.5	77.4
RH chamber [%] corrected	56.9	57.2	47.3	45.5	56.3	55.3	51.7	45.2
$p_v$ Megacup [Pa] corrected	845	1165	1210	1250	1178	1378	1268	1153
$p_v$ chamber [Pa] corrected	1409	1416	1186	1120	1411	1385	1263	1133
$p_v$ difference [Pa] corrected	-564	-252	24	131	-233	-7	5	20
measured flow [ $10^{-8}$ kg/m <sup>2</sup> s]	-47.25	-21.09	2.94	10.94	-28.93	-0.89	0.39	2.48
isothermal flow [ $10^{-8}$ kg/m <sup>2</sup> s]	-38.40	-15.80	0.06	11.20	-21.50	0.26	-0.07	0.08

Values in regular font are taken from the original article; values in italic have been calculated in this paper.

Original measurement values are taken from Appendix B of [29]. The given Megacup and chamber temperatures and relative humidities are actually surface values, so surface resistances do not need to be accounted for.

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1. Temperature profile, vapour pressure profile and vapour diffusion flow for application example, without and with thermal diffusion.

**Table 1:**

Temperature profile, vapour pressure profile and vapour diffusion flow for the application example, without and with thermal diffusion.

	tempe- rature [°C]	vapour pressure [Pa] $\delta_T = 0$	vapour pressure [Pa] $\delta_T = -\delta_p$
interior environment	20.00	1200.0	1200.0
interface interior – concrete	18.60	1199.5	1198.1
interface concrete – insulation	17.77	210.0	237.4
interface insulation – stucco	-9.21	203.4	204.1
interface stucco – exterior	-9.57	200.1	200.5
exterior environment	-10.00	200.0	200.0
		$j_v$ [kg/m <sup>2</sup> ·s] 9.90 10 <sup>-9</sup>	$j_v$ [kg/m <sup>2</sup> ·s] 9.60 10 <sup>-9</sup>

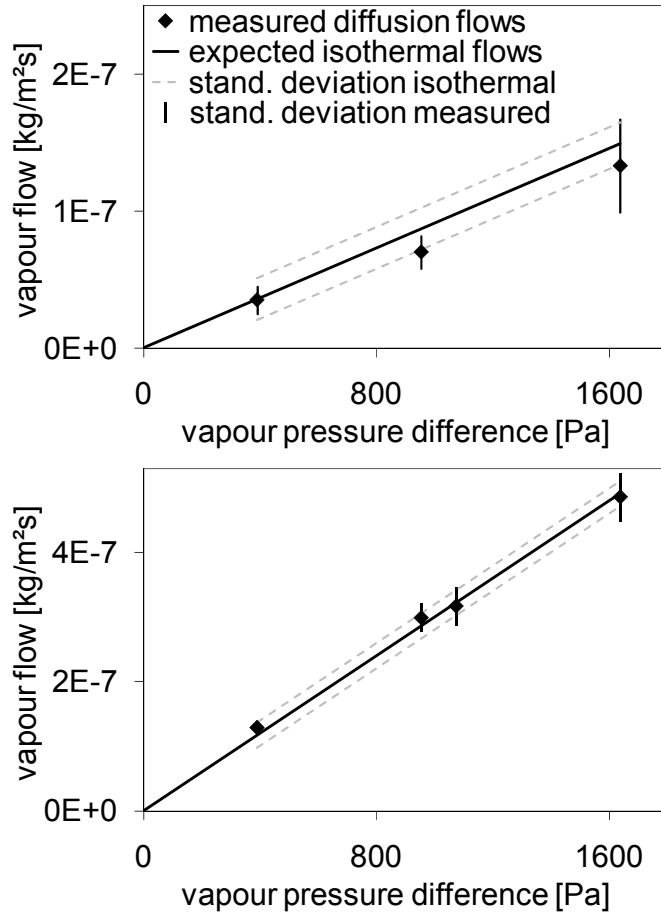


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1. Measured non-isothermal and expected isothermal vapour diffusion flows, and the respective standard deviations, for polystyrene insulation (top) and particle board (bottom).
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9. Corrected non-isothermal and expected isothermal vapour diffusion flows for glass fibre insulation (top) and mineral fibre insulation (bottom).
10. Overview of all original relations (top) and corrected relations (bottom) for the 6 materials as measured on in [8].

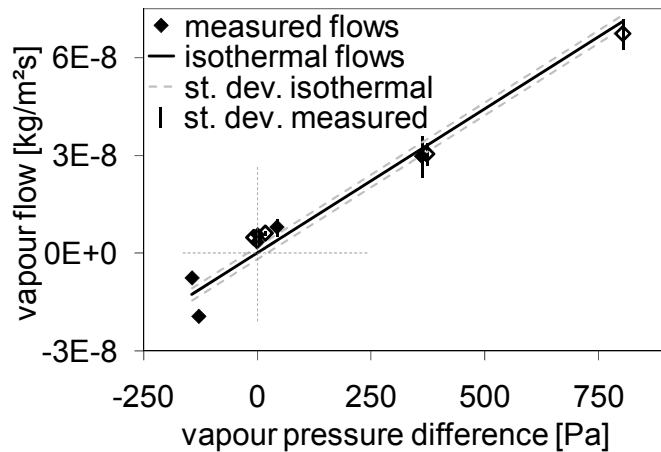
**Figure 1:**

Measured non-isothermal and expected isothermal vapour diffusion flows, and the respective standard deviations, for polystyrene insulation (top) and particle board (bottom).



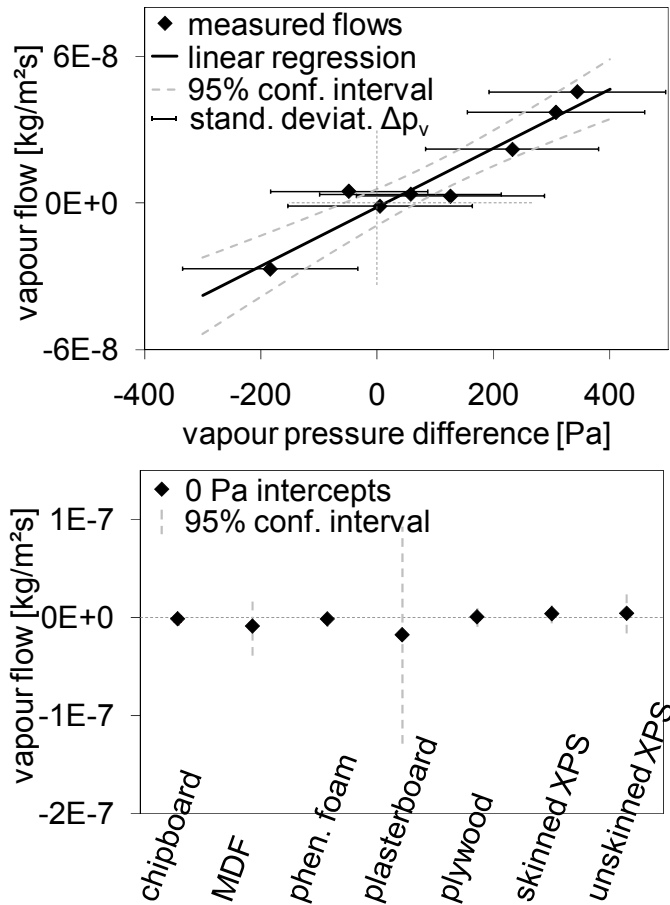
**Figure 2:**

Measured non-isothermal and expected isothermal vapour diffusion flows, and the respective standard deviations, for oriented strand board. The full and hollow markers represent 5 K and 10 K temperature differences respectively.



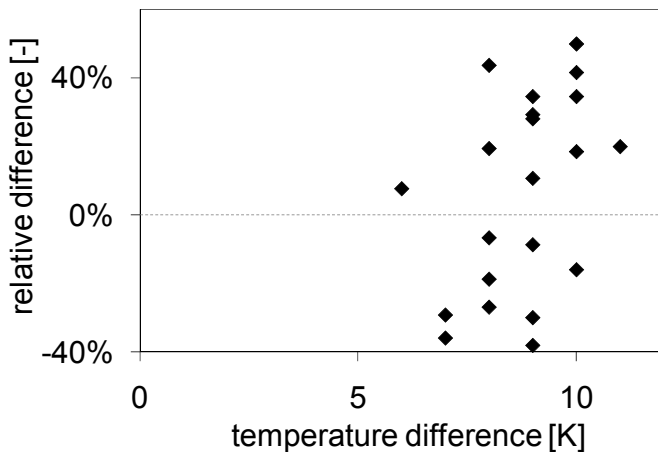
**Figure 3:**

Top: measured non-isothermal vapour diffusion flows and the linear regression through those. The 95% confidence intervals for the regression and the standard deviations on the vapour pressure differences are additionally shown. Bottom: overview of estimated 0 Pa intercepts and the related 95% confidence intervals. Reproduced from [12].



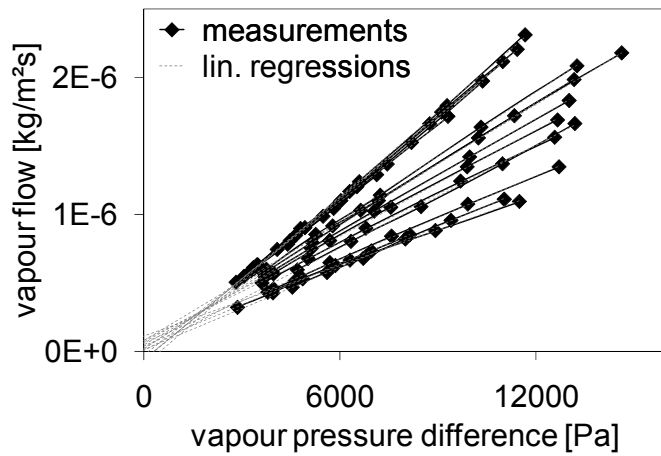
**Figure 4:**

Relative deviations between non-isothermal and isothermal vapour permeabilities, for the 9 materials as measured on in [12].



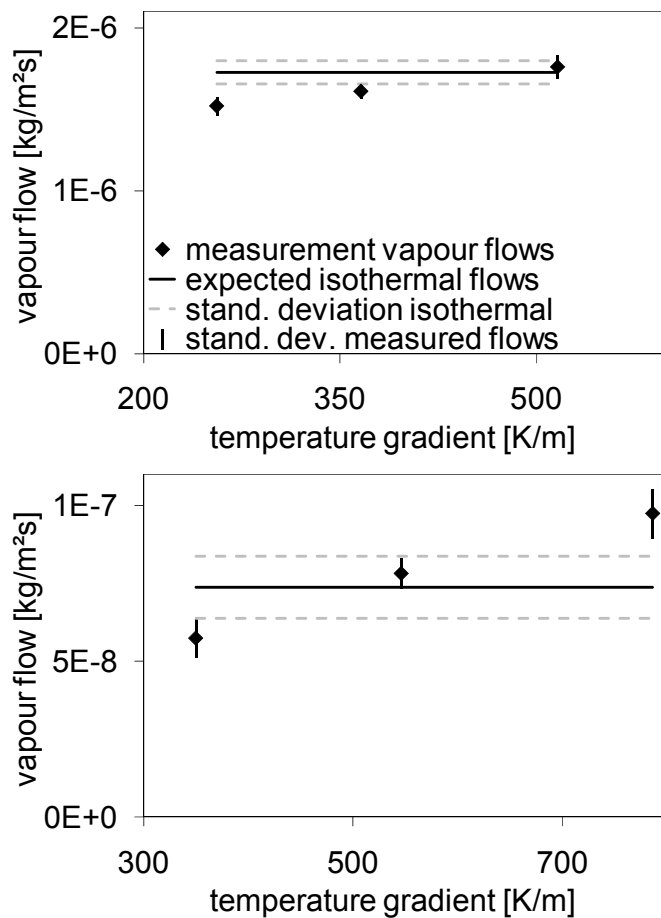
**Figure 5**

Overview of measured vapour flows and their linear regressions for all of Kumaran's measurements.



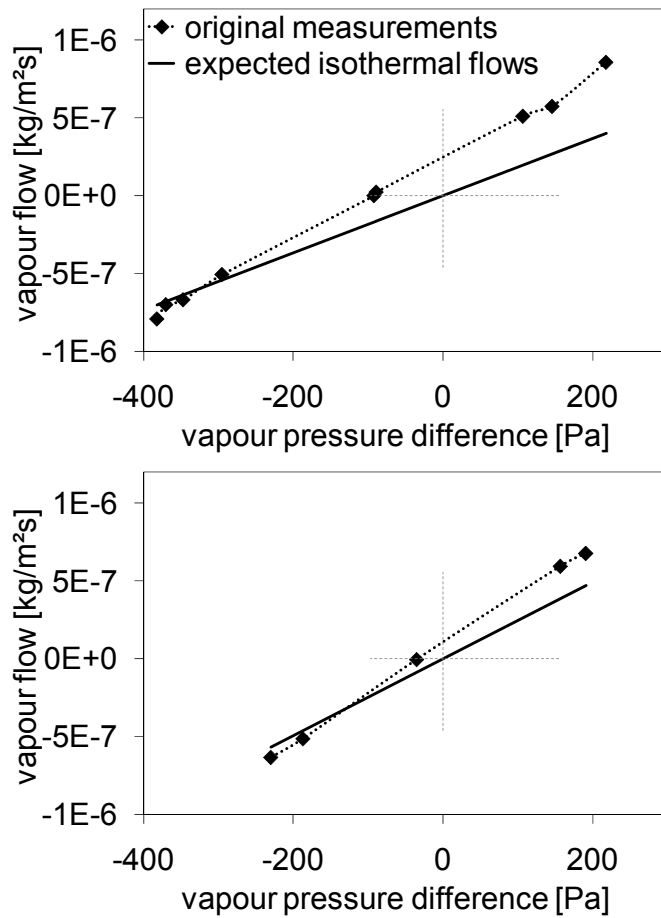
**Figure 6**

Measured non-isothermal and expected isothermal vapour diffusion flows, and the respective standard deviations, for plasterboard (top) and polystyrene insulation (bottom).



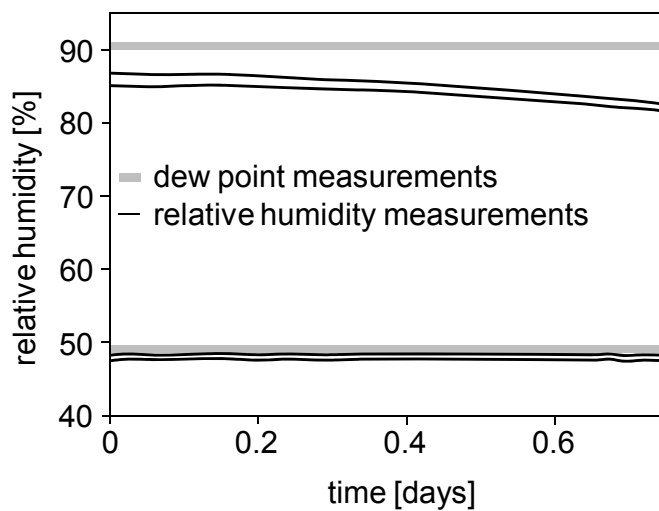
**Figure 7**

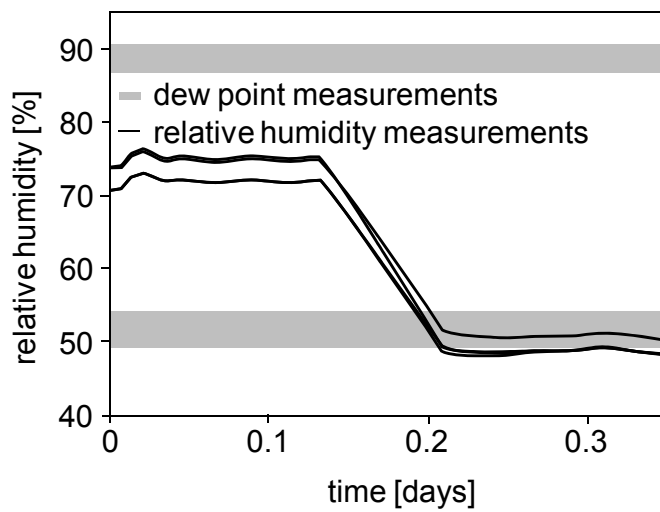
Measured non-isothermal and expected isothermal vapour diffusion flows for glass fibre insulation (top) and mineral fibre insulation (bottom).



**Figure 8**

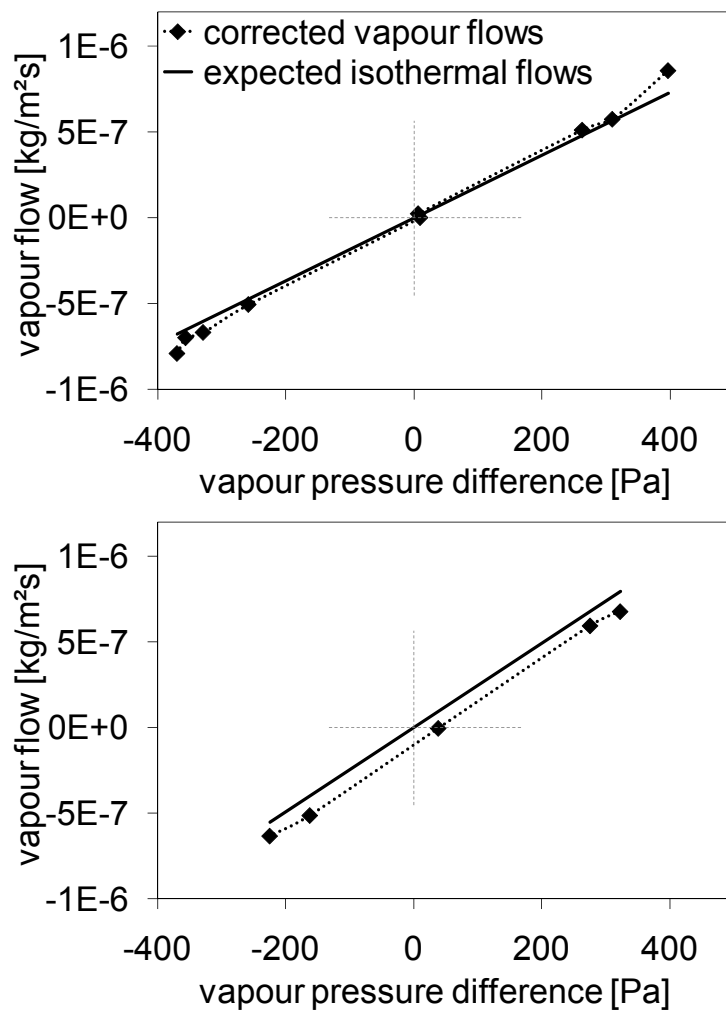
Confrontation of relative humidity sensor measurements with dew point sensor measurements, reproduced from [29].





**Figure 9**

Corrected non-isothermal and expected isothermal vapour diffusion flows for glass fibre insulation (top) and mineral fibre insulation (bottom).



**Figure 10**

Overview of all original relations (top) and corrected relations (bottom) for the 6 materials as measured on in [8].

